Effects of La non-magnetic impurities on the spin resonance of $CeCoIn_5$

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Abstract

The influence of La non magnetic impurities on the spin dynamics of $CeCoIn_5$ was studied by inelastic neutron scattering. In La-substituted systems, the spin resonance peak (observed at $\Omega_{res} = 0.55meV$ in the pure system) is shifted to lower energies but the ratio Ω_{res}/k_BT_c remains unchanged. The excitation broadens till it reaches 0.3 meV equal to the value of the quasi-elastic signal in the normal state. The evolution of La substitution is compared with the evolution of the magnetic resonance in Ni and Zn substituted $YBa_2Cu_3O_7$.

Superconductivity is a macroscopic quantum state resulting from the condensation of electrons in Cooper pairs. In the case of conventionnal superconducvity the pairing mechanism is the weak electron-phonon interaction. Howewer in strongly correlated systems exhibiting a superconducting (SC) state the pairing mechanism is supposed to be of another nature. Examples of such unconventionnal superconductors are the hight temperature superconductors cuprates (HTSC), the heavy fermions compounds (HF) and the new iron-based superconductors. In these compounds, the origin of the pairing is strongly suspected to be the magnetic interaction and a seminal study of the magnetic excitation spectra by inelastic neutron scattering (INS) in the cuprate $YBa_2Cu_3O_{6+x}$ showed the appearence of a sharp excitation called magnetic spin resonance [1] in the SC state. Thus this was then generalized to other cuprates. Such a feedback of superconductivity on the magnetic excitation spectra was backed up by theories of a pairing mechanism of magnetic origins. The recent discovery of similar excitations in HF superconductors UPd_2Al_3[2] $CeCoIn_5$[3] and $CeCu_2Si_2$[4] as well as the new iron superconductors [5] suggests that the magnetic resonance could be a universal feature of the unconventionnal superconductors.

Among HF superconductors the compound $CeCoIn_5$ has the highest critical temperature, $T_c = 2.3K$. It crystallizes in the tetragonal space group *P4/mmm* and can be described as composed alternating $CeIn_3$ and CoIn layers. A quasi-2D nature is supported by de Haas van Alphen, which etablished a Fermi surface composed by nearly cylindrical sheets [6]. As concern the low energy magnetic excitations measured by INS, a quasielastic signal is measured above T_c with a linewidth of 0.3 meV. Below T_c, the spectral shape switches from a quasielastic to a sharp inelastic peak which appears for an energy $\Omega_{res} \approx 0.55$ meV ($\approx 2.7k_BT_c$) at the antiferromagnetic position $Q = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ [3].

The introduction of non magnetic impurities is a useful probe to investigate the microscopic nature of the SC state [7]. In $CeCoIn_5$ this can be achieved by La substitution. Contrary to other types of substitution (Nd on Ce site,Cd on In cite, etc), La does not induce magnetic order but reduces only the critical temperature T_c by $(-0.056T_c)/(1\ \%$ of La substitution) [9][10].

In this Letter we report INS experiments perfomed on single crystal samples of $Ce_{1-x}La_xCoIn_5$ for x=0,0.2,0.35 and 0.5. We found that the ratio between the resonance energy Ω_{res} and T_c remains constant when increasing the La-concentration, whereas the excitation lineshape broadens.

Single crystal samples were grown by the self flux method. Specific heat measurement performed using a commercial Physical Properties Measurement System (PPMS) down to 400 mK, are shown in the inset of Fig. 1. From these measurement we deduce the T_c of each La-substituted system studied in this Letter. Without substitution, T_c is 2.3 K as previously reported [9], for x = 0.02, 0.035 and 0.05, T_c is 1.9K, 1.7K and 1.5K respectively . The specific heat measurements are in agreement with the result obtained in Ref [11]. We observe a reduction at the transition of the specific heat jump while increasing La concentration, together with a broadening of the transition and accordingly an increase of the Sommerfeld coefficient γ for T$\rightarrow$0.

The INS experiments were perfomed on the cold neutron triple-axis spectrometers IN14, IN12 at Institut Laue Langevin, Grenoble and 4F2 at Laboratoire Léon Brillouin, Saclay. In the three experiments, the incident beam was provided by a vertically focusing pyrolytic graphite (PG) monochromator (double-monochromator in the case of 4F2). A liquid-nitrogen-cooled Be filter was placed before the sample in order to avoid any higher order contaminations. Measurements were perfomed with a fixed final wavevector $k_f = 1.3\,\text{Å}^{-1}$ for IN14 and IN12, and $k_f = 1.35\,\text{Å}^{-1}$ for 4F2. The collimations were 60'-open-open. The energy resolution determined from the full width at half-maximum (FWHM) of the incoherent signal was 0.12 meV on IN14, 0.10 on IN12 and 0.15 meV on 4F2. The different samples consisted of assemblies of about 30 single crystals of $Ce_{1-x}La_xCoIn_5$ co-aligned and glued with Fomblin oil on two thin aluminum plates. The mosaic spread of the three assemblies, as measured

on a rocking curve through the (1,1,1) Bragg reflection, extend from 1 degree to 1.5 degree. The sample was put in a ^{3}He insert for the experiments on IN14 and IN12, and in a dilution insert on 4F2 with [1,1,0] and [0,0,1] defining the scattering plane.
The measured neutron intensity without the background is proportionnal to the scattering funcion S(**Q**,E) itself related to the imaginary part of susceptibility $\chi''(\mathbf{Q}, E)$.

$$S(\mathbf{Q}, E) \;=\; n(E,T)\,\chi''(\mathbf{Q}, E)$$

χ"was analysed using an "inelastic Lorentzian" spectral function:

$$\chi''(\mathbf{Q}, E) \;=\; \frac{1}{2}\left[\frac{\chi_Q \Gamma_Q E}{(E-\Omega_{res})^2+\Gamma_Q^2} + \frac{\chi_Q \Gamma_Q E}{(E+\Omega_{res})^2+\Gamma_Q^2}\right]$$

$n(E,T) = 1/(1-e^{-E/k_BT})$ is the detailed balance factor, Γ_Q is the relaxation rate, Ω_{res} is the resonance energy and χ_Q is susceptbility at the wave-vector Q. All the energy spectra were taken at $\mathbf{Q} = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ except the background measurements.
The magnetic excitation spectra measured at $Q = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ as a function of La-substitution is shown on the figure 1. The data for x=0 are taken from Ref [12]. The dashed line corresponds to the background signal obtained by performing an energy scan shifted in **Q** position. For the experiment on IN12 and IN14 the background was measured at $\mathbf{Q} = (0.8, 0.8, \frac{1}{2})$ and for 4F2 at **Q** =(0.412,0.412,0.8). Without substitution, the resonance peak is reported in [12] at 0.55 meV with a relaxation rate of 0.07 meV. In 2 % La-substituted system the resonance peak shifts to $\Omega_{res} = 0.45$ meV and endures a substantial broadening reaching a relaxation rate Γ of 0.3 meV. A La-substitution of 3.5% shifts the resonance peak to $\Omega_{res} = 0.35$ meV but Γ remains constant at 0.3±0.05 meV. For this latter concentration, it is worthwhile to note that the resonance peak is no more a well-defined inelastic excitation since $\Omega_{res} \approx \Gamma$. The spectra with a 5% La-substitution (not shown here) presents no more resonance peak. Either the peak is too broad to be resolved or the excitation occurs at too low energy to be separated from the incoherent signal. The parameters extracted from the data fit are summarized in figure 2. In this figure the linear fit for the evolution of the resonance energy Ω_{res} as a function of La-substitution corresponds to a rate of $(-0.058\Omega_{res})$/(1% of La substitution). As concern the relaxation rate, its increase is probably related to the diminution of the specific heat SC jump and the concomittant increase of the Sommerfeld coefficient.

Constant energy scans were perfomed along the a and c-axis around $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ in order to measure the correlation lenghts of the resonance under La-substitution. Figure 3 shows spectra measured at 0.5 meV with a subsitution of

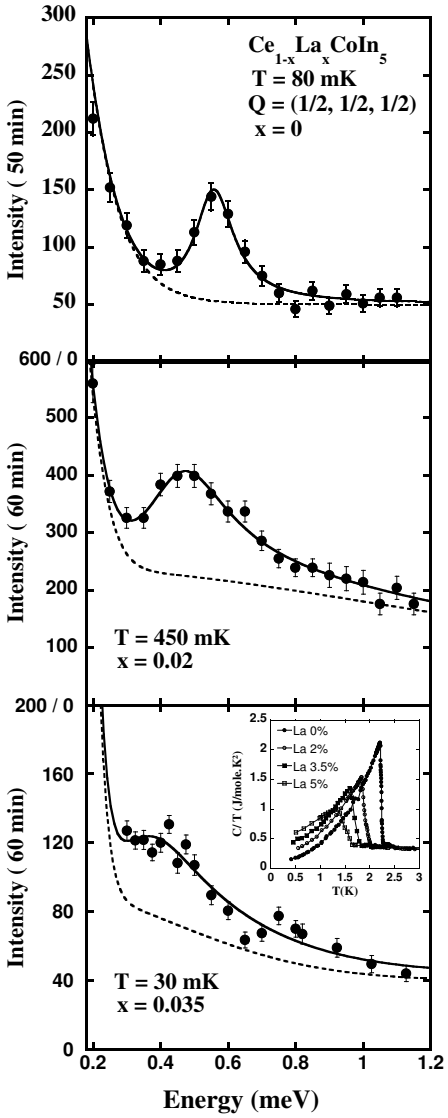

FIG. 1: Excitation spectra measured at $\mathbf{Q} = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ for $Ce_{1-x}La_xCoIn_5$ with x = 0, 0.02 and 0.035. The solid lines are "inelastic Lorentzian" fits and the dashed line indicates the background as described in the text. The inset shows the specific heat(C_p) measurements of La-substitution of 0%,2%,3.5% and 5%

2%. The scans are analyzed with a gaussian lineshape. The background is determined by the measurements at hight temperature where the magnetic spectrum is no longer peaked in **Q**. The signal still peaks at $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ as for a pure compound. The correlation lenght obtained from the inverse of the gaussian half-width at half maximum are $\xi_c = 5.1 \pm 0.1$ Å and $\xi_a = 11.8 \pm 0.6$ Å. In comparison with the pure compound [3], the correlation lengths remain similar in both directions (for a pure compound $\xi_c = 6.5 \pm 0.9$ Å and $\xi_a = 9.6 \pm 1$ Å).
The evolution of the resonance peak of a substituted sys-

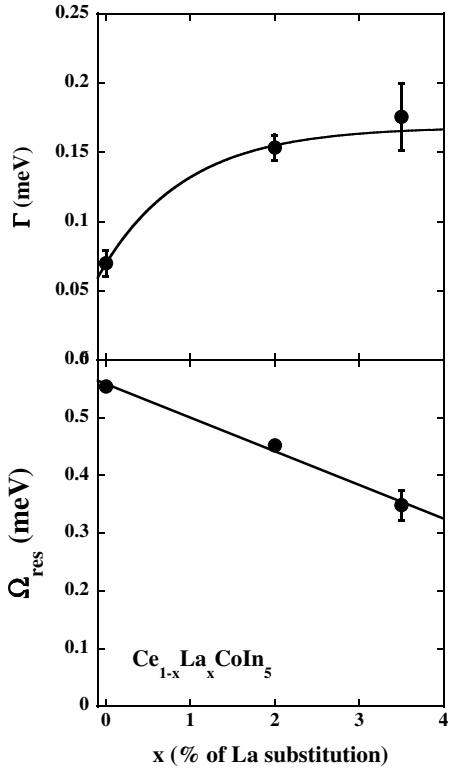

FIG. 2: Evolution of the relaxation rate Γ and the resonance energy Ω_{res} as a function of the La-substitution in $CeCoIn_5$. The lines are a linear fit for the resonance energy and a guide for eye for the relaxation rate

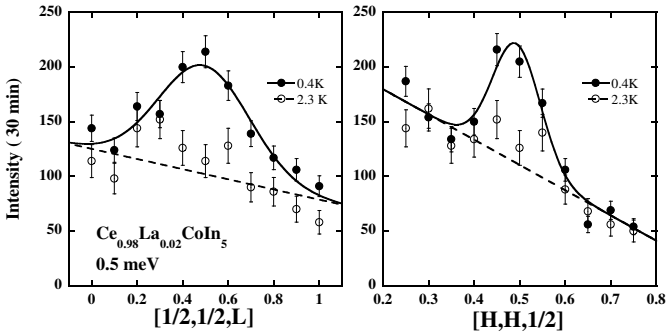

FIG. 3: Excitation spectra measured at E =0.5 meV in the direction $[\frac{1}{2},\frac{1}{2},L]$ and $[H,H,\frac{1}{2}]$ for $Ce_{0.98}La_{0.02}CoIn_5$. The solid line is a Gaussian fit and the dashed line indicates the background as described in the text.

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tem was studied as a function of temperature. Indeed in the pure compound the resonance peak has been observed only in the SC state [3] showing the strong coupling between the resonance excitation and superconductivity. On figure 4 we report the evolution of the $\Omega_{res}/\Omega_{res}(T=0)$ as a function of T/T_c for a pure compound 0% (extracted from the ref[3]) and a La-substitution of 2%. The evolution in temperature of Ω_{res} for a 2% La-substituted system matches the evolution of a pure compound. The presence of impurities seems to have no influence on the coupling between superconductivity and resonance excitation since there is no persistance of the resonance above T_c.

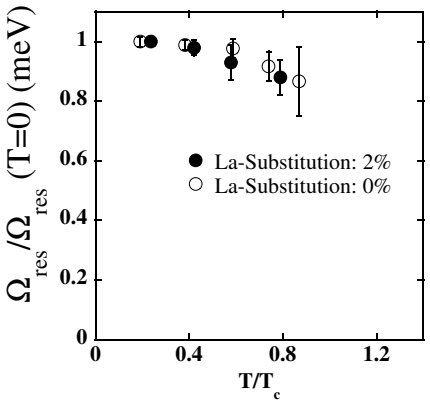

FIG. 4: Evolution of the resonance energy Ω_{res} of $Ce_{0.98}La_{0.02}CoIn_5$ and $CeCoIn_5$ from [3] at $\mathbf{Q}=(\frac{1}{2},\frac{1}{2},\frac{1}{2})$ as function of the ratio T/T_c with for a pure compound T_c= 2.3K and for a 2%-substituted compound T_c= 1.9K

When we compare the shift of Ω_{res} and the decrease of T_c over the studied compounds, we obtain the ratio $\Omega_{res}/k_BT_c \approx 2.7$. Such a constant ratio between T_c and Ω_{res} has already been observed in other HTSC ($YBa_2Cu_3O_7$ [13], $Bi_2Sr_2CaCu_2O_{8+x}$ [14]) where the ratio is about 5.1. The linear relation in the HTSC has been established by changing the oxygen doping and so far the number of carriers but the La-substitution in $CeCoIn_5$ corresponds at first approximation more to the insertion of non-magnetic impurities. There are few models which have been developped concerning the effect of non-magnetic impurities on the resonance for a d-wave superconductor. To our knowledge there is only one study which describes precisely this effect,showing a decrease of the resonance energy and an increase of the resonance width [16] upon the introduction of impurities , in agreement with our results even if this theory was developped using a band structure adequate for the cuprate.
Two INS experiments have been performed on the system YBCO with the substitution of Cu by Zn non-magnetic impurities and Ni magnetic impurities [13] [15].If we make an quantitative comparison with these two cases, some features drawn attention: the Zn-substitution and the Ni-substitution induce both a T_c reduction but the Zn-one has a higher rate of suppression of T_c : $(\approx -0.13T_c)$/(1% of Zn substitution) in comparison of $(\approx -0.04T_c)$/(1% of Ni substitution) [15]. The influence of both impurities is totally different concerning the evolution of the spin resonance in energy and in temperature. The 1% Zn-substitution does not shift Ω_{res} and so increases the ratio Ω_{res}/k_BT_c in comparison of a pure $YBa_2Cu_3O_7$ compound. Though the 3% Ni-substitution

leads to a decrease of Ω_{res} while conserving the ratio Ω_{res}/k_BT_c=5.1. As concern the width of the resonance peak in energy, no noticeable increase of the relaxation rate is observed at low values of substitution [15] in $YBa_2Cu_3O_7$ with both Zn and Ni substitution contrary to our case. But the large value of the relaxation rate in $YBa_2Cu_3O_7$ could hide a small augmentation of the width. Moreover the magnetic signal in the 1% Zn-substituted compound does no disappear at T_c but half of the integrated intensity remains above T_c. As concern the Ni-case, the magnetic signal vanished upon increasing the temperature up to T_c as the La-substituted $CeCoIn_5$ compound. In conclusion the case of La-impurities in $CeCoIn_5$ is closer to the case of Ni magnetic impurities than Zn non-magnetic impurities in $YBa_2Cu_3O_7$.

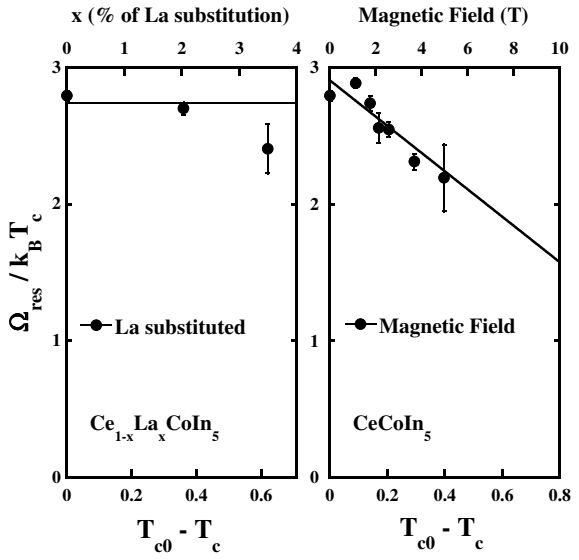

FIG. 5: Evolution of the resonance energy Ω_{res} divided by the critical temperature as function of T_{c0}-T_c controlled by La-subsitution(left panel) or magnetic field (right panel with T_{c0}=T_c for pure compound without magnetic field

Finally we compare the La-substitution with another way to tune the superconductivity, the application of a magnetic field. If we consider that the influence of the magnetic field is just a diminution of the SC gap and thus the critical temperature, the evolution of the resonance as a function of critical temperature for the La-substitution experiment (figure 5) and for magnetic field from Ref.[12] shall be the same. The figure 5 proves that magnetic field has a different effect on the spin resonance that the La-substitution for which the ratio between the resonance and the critical temperature approximately remains constants with $\Omega_{res}/k_BT_c \approx 2.7$ while the application of a magnetic field diminishes this ratio. An explanation of this decrease would be to introduce a Zeeman splitting of the spin resonance under magnetic field. In this case, the lowest energy mode decreases with a linear rate while the magnetic field increases as shown in Ref. [12]. This behaviour is consistent with the most common models for magnetic resonance that may apply to $CeCoIn_5$: the spin triplet exciton or the magnon model [17]. As discussed in Ref.[12], a definitive proof would be to clearly observe another mode of the multiplet at higher energy but the comparison between the La-substitution and the magnetic field effect, that span a similar range of T_c, suggests indeed a multiplet nature of the spin resonance in $CeCoIn_5$.

The accurate study of the resonance as a function of La-substitution in $CeCoIn_5$ shows a almost constant ratio $\Omega_{res}/k_BT_c \approx 2.7$ with a broadening of the excitation. Theses observations are in agremment with a theoretical model developped for $YBa_2Cu_3O_7$ but our results are in stark contrasts with the INS experiments perfomed on $YBa_2Cu_3O_7$ with Zn non-magnetic impurities. Theses overall issues deserve further theoritical studies.

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